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The instant invention concerns a reactor and a method for execution heterogeneous-catalytic gaseous phase reactions.

Examples for such heterogeneous-catalytic gaseous phase reactions are for instance the preparation of ethylene oxide from ethylene and oxygen, the preparation of acrylic acid from acrolein and oxygen, the preparation of methyl acrylic acid from methacrolein and oxygen, the preparation of styrene by dehydration of Methylphenylcarbinol, the preparation of methanol from synthesis gas, the Methanisierung of synthesis gas, the conversion of very low-temperature, the conversion of water with olefines to alcohols, the Oxychlorierung of olefines or aromatics, or the preparation of methylamines from methanol and ammonia. The cited examples represent however in keinsten manner a complete enumerating of all according to invention conversions feasible with the reactor and method.

Reactor types for execution heterogeneous-catalytic gaseous phase reactions are in large number known. In the predominant plurality of all technical applied heterogeneous-catalytic gaseous phase reactions a Geradrohrbündelreaktor becomes used. Such reactors are for a long time state of the art and become from numerous firms prepared and commercial sold. The resulting waste heat becomes used in Geradrohrbündelreaktoren by a suitable cooling medium, like usually for the example pressure water or high-boiling hydrocarbons, discharged and steam production.

This method, in particular the Geradrohrbündelreaktor used thereby, some disadvantages are immanent. So the length of the catalyst pouring and thus the reactor for a satisfactory conversion relative large, in the rule, gone through by the gas mixture, must be some metres. This leads to high costs for the gas compression, since the pressure drop is usually significant over the reactor-prolonged. An other disadvantage of this type of reactor is that the capacity of such bank of tubes reactors is limited. The reactors consist of up to several thousand single tubes, which are in a reactor body summarized. An increase of the production achievement of such a reactor is not possible by magnification of the diameter of the single tubes, since in this case the average distance of the single catalyst particles of the cooled or heated pipe wall becomes larger. Thus the cooling becomes poorer, so that single, superheated, catalyst particle as ignition source for "going through" the reaction to function to be able. By the high heat of reaction freed in such a case the catalyst becomes usually irreversible damaged. An increase of the production achievement of a bank of tubes reactor is thus only possible by increase of the tubing's number with suitable diameter of the single pipes. Also this procedure leads however to problems. Since which must orient required crush strength of the reactor body at the high pressure of the coolant and not at the smaller pressure of the reaction gas mixture, these reactors are, expensive made with exception of salt bath-cooled reactors for reactions with temperatures over 300 DEG C, very thick walled, heavy, and concomitantly expensively and only bottom very large difficulties transportable. The nowadays realized sizes reach already the boundary of the economic meaningful. An other major disadvantage of this type is that the Geradrohrbündel is very susceptible to thermal stresses, which become caused by temperature differences between reactor pressure vessel wall and reaction tube sheet. Thereby the danger exists the fact that with inappropriate operation or serious operational disturbances reaction tubes break or peels off or that breaks the reactor pressure vessel wall.

In principle different type of reactor for execution heterogeneous-catalytic gaseous phase reactions is based to another suitable type of the heat dissipation on the application of a loose catalyst pouring with embedded heat exchanger or, in case of exothermic reactions. The state of the art large is laid down and in numerous writings also here. Thus about US A2 stress 744,813 a reactor with loose catalyst pouring, straight cooling tubes and axial gas stream, EP-B 1 82 609 (ICI) a similar reactor with radial gas stream. Reactors with straight cooling tubes in a loose catalyst pouring need however planar tubesheets and suffer thereby from the same problems as a conventional Geradrohrbündelreaktor. Beyond that designed itself the filled and deflation of the catalyst substantial more difficult. Another type of reactors with loose catalyst pouring and embedded heat exchanger an used heat exchanger from layers of coiled tubes (DE 34 14 717 (lime tree), DE 28 48 014 (lime tree), DE 39 35 030 (lime tree)). With coiled tubes the need of planar tubesheets and problems with thermovoltages are not void are due to the elasticity tubingwind practical given. The tubes in this type crosswise, by the special geometry of the tube assembly are the gas mixture and thus the convective heat transport greatly increased are flowed against. Beyond that the average distance of a catalyst grain of the next heat exchange-flat is smaller than in comparable Geradrohrbündelreaktoren. Beide of effects leads to a significant reduction the required heat exchange-flat opposite Geradrohrbündelreaktoren of the same catalyst volume on for instance the half. The specific heat exchange-flat can become the other to the reaction progress adapted or the temperature of the flowing of the coolant in the tubes can varied become (EP 339,748 a2 (Shell)). An other heat transfer possibility exists in adding reactants or inert gases along the catalyst pouring (DE 28 48 014, DE 39 35 030) or, with exothermic reactions, in the Durchleiten of of a cold reactor entrance gas by the catalyst pouring (the USA 1,835,827 (you Pont)).

One with practical each heterogeneous-catalytic gaseous phase reaction appearance which can be observed is that the value product of the reaction becomes at least partial decomposed by contact with the catalyst or bottom reaction conditions dominant used to its synthesis in the catalyst bed again, so that a part of the product in the rear part,

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formed in the front part of the catalyst pouring, is again destroyed. As was now found, this retention time effect for the yield is more significant as for instance a moderate temperature rise contrary to the predominant opinion. With exothermic decomposition of the value product, for instance the other oxidation of the product of a selective oxidation with oxygen to carbon dioxide, the catalyst can become irreversible damaged by going through the reaction bottom excessive heat development. With other reactions the catalyst can become by decomposition of the value product bottom coke separation passivated. Another, disturbing influence of value product present in the reaction gas mixture can be the inhibition of the desired reaction by the value product. In everyone of the shown cases it is of advantage to remove the value product as rapid as possible from the catalyst bed.

An obvious procedure to moderate the shown above problem of the decomposition of the value product with the passage by the catalyst pouring is, to hold the catalyst distance gone through by the reaction gas mixture as short as possible. The conversion at feeds to the value product sinks thereby however which mostly by temperature rise compensated can become, if this temperature rise does not have negative effects on the reaction happening. Beyond that the pressure drop over the reactor and thus the compression costs can become reduced by a shortening of the catalyst pouring, or, alternative, the production achievement of the reactor can become by magnification of the reaction gas quantity gleiteten per unit time by the catalyst elevated. With processes, which work after the principle of the circle gas driving fashion with a continuous removing from products, the pressure drop can become by a shortening of the catalyst layer even so far minimized that such an increase of the reaction gas quantity led by the catalyst pouring is possible that with constant production at value product of the turnover degrees per reactor passage reduced can become. This brings usually the additional advantage to an improved selectivity, so that for a constant production held fewer starting material used must become than with conventional operation. However secured must be with elevated gas throughput and reduced turnover degree that Aufarbeitungs-und purification system can master the resulting amounts of gas. The optimization of reactors and processes in the parameter field outlined here is the conventional object of a person skilled in the art on this field.

Object of the instant invention is it to find an improved process for execution heterogeneous-catalytic gaseous phase reactions as well as a reactor for the execution of the reactions.

Improved processes for the execution of such reactions, as for instance the preparation of ethylene oxide from ethylene and oxygen, the preparation of acrylic acid from acrolein and oxygen, the preparation of methacrylic acid from methacrolein and oxygen, the preparation of styrene by dehydration of Methylphenylcarbinol, the preparation of methanol from synthesis gas, the Methanisierung of synthesis gas, the conversion of very low-temperature, the conversion of water with olefines to alcohols, the Oxychlorierung of olefines or aromatics or the preparation of methylamines from methanol and ammonia possess large economic importance.

This object becomes 1 dissolved by the characterizing features of the claim.

Other features of the invention process are subject-matter of the Unteransprüche 2 - 10.

In the claims 11 - 20 is methods to the preparation of value products by means of the shown reactor and method according to invention shown.

A shortening can particularly favourable be reached from the reaction gas mixture to continuous catalyst pouring by the fact that the reaction gas mixture does not flow by shortened catalyst-filled tubes of a reactor of conventional type, but that a loose catalyst pouring becomes applied, those by the reaction gas mixture not in more axial, but in radial direction is flowed through. The direction of flow can be thereby centrifugal or centripetal, whereby the centripetal direction is particularly favourable whole, since the flow rate of the gas in this case of the entry increases into the reactor to the exit from the reactor continuous. The residence time of the reaction gas mixture in a certain layer thickness of the catalyst pouring sinks therefore with Reaktionsfortschritt. Dadurch already formed value product accelerated exploit-decreasing post-reactions eventual remote from the catalyst bed and is extracted. The case of a centrifugal current of the reaction gas mixture can exhibit bottom special circumstances however advantages over a centripetal current, in particular with strong volume-increasing reactions. With such reactions a retention time optimization can become conducted by a suitable dimensioning of the reactor or by other measures, for instance the incorporation of Verdrängerkörpern at the outer edge of the reactor, also with centrifugal stream.

That whole particular advantage of the reactor and method according to invention in its normally preferable embodiment is however the accelerated discharge of the value product of the catalyst bed with centripetal gas stream. To the negative effects to large residence times of the value product in the catalyst bed, for the example yield loss by its decomposition bottom possible damage of the catalyst, approximately by temperature rises with total oxidation of the value product of a selective oxidation with oxygen or coking of the catalyst, with the reactor and method according to invention just as effective one meets as the passivation of the catalyst by formed value product. As mentioned already above became, the effect of a too prolonged residence time is possible on the yield at value products significant Reaktionstemperatur. Dadurch elevated as the effect is it to raise with unsatisfactory turnover degrees the reaction temperature by pyrometric measures with reaction progress and to complete so the conversion.

An embodiment of the invention with the substantial inventive features is in the drawing shown and becomes in the following more near described.

Fig 1 shows a cross section by the reactor according to invention,

Fig 2 shows a cross section by the reactor according to invention with additional controllable heating circle circulation.

It is absolutely required to exhaust the resultant heat of reaction. Here the use of a radial current reactor is particularly favourable, is incorporated with which into the catalyst pouring (01) a bank of tubes heat exchanger (02) with coiled cooling tubes (03). Into these a suitable heat distribution medium medium flows, for the example pressure water or kerosene. In a possible and favourable configuration the tube bundle consists of several concentric tubing situations, which proceed from the hemisphere soil of the distributor (04) and become at the likewise hemispheric soil of the collecting tank (05) again combined. Number, distance in more horizontal and vertical direction as well as size and direction of the slope of the single windings can become varied and thus the pyrometric requirements and the size

of the catalyst particle adapted. During centripetal flow the reaction mixture steps the catalyst pouring over several nozzles (06) into the outer concentric distributor annular space (07) of the reactor, which becomes from the receiver wall (08) and one with these connected circumferential shirt (09) formed.

This shirt separates the annular space from the catalyst pouring. The reaction gas mixture partitioned and arrives itself by bores into the catalyst pouring. This flows through it in radial direction up to the inner collecting pipe (10), which likewise possesses passage openings. The production gas mixture over discharge part (11) is led out from here from the reactor. If a reverse direction of flow is to become realized, then this with the same structure possible is.

Likewise a configuration is possible, used with which a part of the heated cooling medium (for the example in the form of water vapor) becomes the heating of cold reactor entrance gas. For this or the several concentric windings (12) of the cooling circuit - coiled cooling tubes (03) - can do a disconnected and to of recycled, heated cooling medium flow through heating cycle combined to become (13). This is with centripetal, in addition, possible with corresponding arrangement of the disconnected windings with centrifugal direction of flow.

The reactor with an heat exchanger with coiled tubes possesses significant better pyrometric properties as the conventional type also prolonged-flowed through Geradrohrbündel. Thus it results as advantage that the reactor can become significant compact constructed during same production achievement and dimensions, weight and capital outlays substantially reduced become. As other advantage it is to be called that possible with this type of building an increase of the production achievement is by magnification of the reactor, while with the nowadays realized sizes with Geradrohrbündeln the boundary of the economic meaningful achieved is. Finally the constructive structure with coiled tube bundle is more favourable regarding arising thermal stresses, which become caused by temperature differences between reaction gas mixture and cooling medium, thus between reactor pressure vessel wall and condenser tube wall. Large temperature differences can be borne by the elastic feather/spring behavior of the coiled tubes, without causing mechanical stresses, which for the normal operation, but particularly for the safe control of operational disturbances is favourable. The reactor and method according to invention is thus an additional not insignificant increase of the working reliability opposite the state of the art to own.

An other possibility to the discharge of the heat of reaction in the reactor and method, according to invention resulting with exothermic reactions, exists in its direct use to the heating of the cold reaction gas mixture by the method of the so called fresh gas feed. The number and the layer of the fresh gas discharge positions in the catalyst pouring can be thereby depending upon amounts of heat which can be exhausted resultant at the given locus in the catalyst pouring and different. By this method by active cooling or becomes reduced over a separate heat exchanger from the reactor amount of heat which can be exhausted. The technical mechanisms necessary for an active cooling or a separate heat exchanger can become therefore small dimensioned and the costs resulting with its preparation and assembly are smaller.

Likewise with exothermic reactions more applicable a combination that is managing described cool methods, which becomes used in a reactor with radial current of the reaction gas mixture by the catalyst bed. Warm one, which does not become consumed by the introduction of cold reactor entrance gas, becomes discharged thereby by a cooling medium flowing in coiled tubes by the catalyst bed.

The invention process for the execution of a heterogeneous-catalytic gaseous phase reaction can become both in circle gas driving fashion and with unique passage of the reaction gas mixture by the catalyst bed conducted. In the circle gas driving fashion the reaction gas mixture in the circuit will become by the catalyst-filled reactor sent, formed products as well as developed byproducts and fabrics, which would enrich themselves in the circle gas otherwise, by suitable means from the circuit remote and the appropriate amounts at feeds as well as if necessary reaction modifiers the gas mixture before renewed passage by the reactor admixed. With the embodiment with unique gas-depressed through the reactor the entire gas mixture is regenerated after the reactor.

Exactly the same it is according to invention possible with the reactor and method to accomplish endothermic reactions. In the tubes of the heat exchanger then an heating medium, the described above favourable pyrometric properties of the reactor according to invention flows applies to an heat flow in reverse direction, how it becomes required with endothermic reactions, just like for exothermic reactions.